

Study of the hidden-order of URu₂Si₂ by point contact tunnel junctions.

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URu₂Si₂ presents superconductivity at temperatures below 1.5 K, and a hidden order (HO) at about 17.5 K, both electronic phenomena are influenced by Fano and Kondo resonances. At 17.5 K the HO was related in the past to a Peierls distortion that produces an energy gap deformed by the resonances. This order has been studied by more than 20 years, still without a clear understanding. In this work we studied the electronic characteristics of URu₂Si₂ in a single crystal, with tunneling and metallic point contact spectroscopies. In the superconducting state, we determined the energy gap, which shows the influence of the Fano and Kondo resonances. At temperatures where HO is observed, the tunnel junctions spectra show the influence of the two resonances. Tunnel junctions characteristics show that the Fermi surface nesting depends on the crystallographic direction.

I. INTRODUCTION

URu₂Si₂, is one of the most studied materials, after more than 20 years the electronic characteristics are still not well understood. One notable example is the so called hidden order which remains unexplained. Transport measurements in this compound have shown many details related with the general behavior; from room temperature to 70 K URu₂Si₂ presents a characteristic typical of a Kondo system associated to the interaction of heavy f electrons and spd conducting electrons. With a Kondo lattice formed at $T_K \sim 70$ K [1, 2], below this temperature the resistivity dramatically drops, and at $T = 17.5$ K an anomaly is observed. In the past this was related to a spin density wave (SDW). However this feature is called the Hidden Order (HO). It presents many diverse electronic characteristics that modifies other occurring processes [3–6]. At lower temperature below ~ 1.5 K the compound, turns into a superconducting state which many researchers believe must be anomalous in the sense that does not follow the BCS model [7–12].

As mentioned before the HO was associated in the past to a Peierls distortion, but careful experiments carried out to detect the Peierls (SDW) distortion did not find any magnetic order, only a quite small staggering magnetism, probably related to magnetic impurities, not related to SDW behavior. This absences imply that the gapping on the Fermi surface may be related to another unknown electronic phenomena [3, 13]. Actually, doubts still persist about the physics behind the anomalies at about 17.5 K. The HO changes transport, and thermal properties, and opens an energy gap, and altering the superconducting behavior [5, 6, 8, 14–17]. The occurrences of the HO, and the relationship with the Fano and Kondo resonances is not completely understood, nobody knows the influence of the two resonances and how those are affecting the general behavior, for instances the manner

that the Kondo lattice at high temperature and the resulting distortion at 17 K modifies the electronic characteristics. Experiments with very sensitive techniques as tunneling and point contact spectroscopy (PCS) provide the observation of the Fano resonance that distorted the feature of the energy gap, at $T \sim 17.5$ K as observed by Schmidt, et al, with Scanning Tunneling Spectroscopy [18].

Many researchers who investigated this transition demonstrated that this is a second order thermodynamic transition [7, 9, 19–21]. The effects of the resonances at the transition show the consequence of a partial electronic screening that modifies the transport and thermal properties because the strong hybridization between spd conducting electrons and heavy localized f electrons [18, 22]. The result of all those processes is that the Kondo and Fano lattices are spatially modulates. The signature of these lattices are displayed as modifications at the Fermi surface. Thus, portions of the Fermi surface at high and low temperatures are distorted and observed with many experimental tests and theoretical studies [1, 7, 23–25].

This work shows our new experimental observations obtained with techniques that give additional information about the features and characteristics of this compound using tunneling and PCS [26–30].

The studies at high temperatures from 40 to 1.7 K were performed using tunnel junctions to observe the HO that develops at ~ 17.5 K. We studied the anomalous features of the gap, using a well characterized URu₂Si₂ single crystal [19, 20, 31], we measured the spectroscopic characteristics of the crystal in two crystallographic directions.

At lower temperature the superconducting state was studied with metallic point contacts (PCS) in the range of temperatures where superconductivity develops, the studies were performed from 2.5 to 0.3 K. PCS junctions were in the diffusive limit but close to the ballistic regime. In all experiments, here reported, from 40 K to 0.3 K the influence of the Fano and Kondo resonances was observed. In the superconducting state the influence of both resonances was clearly evidenced; the feature of the energy gap was completely distorted, and the effect

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of these resonances is clearly noted when examined the evolution with temperature of the energy gap.

In the range of temperatures where HO is set, the influence was studied in two directions of the crystal, the Fano features are different in both directions.

This work reports tunneling and point contact experiments performed in URu_2Si_2 single crystals. At temperatures from 1.7 K to 40 K, tunneling spectroscopy shows the effect of the Fano and Kondo resonances on the energy gap, and the dependence on the crystallographic direction. The energy gap in the a direction is bigger than the gap in the c direction. In the superconducting state, with PCS we found that the energy gap as a function of temperature follows the BCS prediction after the Fano anomaly is subtracted from the differential conductance.

II. EXPERIMENTAL DETAILS

The URu_2Si_2 single crystals used in this study were growth by Czochralski method and annealed for one week at 850°C . They have similar characteristics to that reported elsewhere [19, 20, 31]. The crystals have a platelet-like shape with the crystallographic c axis perpendicular to the plane of the platelet, typical dimension of the used crystals were about $2\times 3\times 0.5\text{ mm}^3$.

The samples were characterized by resistance as a function of temperature measurements. The applied current was $100\text{ }\mu\text{A}$. The resistance ratio between room temperature and low temperature resistance $R(300\text{ K})/R(2\text{ K})=38$. The anomaly associated to the HO is observed clearly at about 17.5 K. At lower temperature, the superconducting transition temperature is at $T_C=1.37\text{ K}$ ($R=0$), this transition is also similar to the observed in other experiments by other researchers. The transition temperature width was 0.15 K. Two type of junctions were formed, tunnel junctions were build by using a tip of Au(W) wire and the URu_2Si_2 crystal. The insulating barrier that forms the tunnel junction was the native oxide on the surface of the compound, and/or the oxide on the tip of the wire. The reason to assume that the junctions in our experiments behaves as tunnel or point contacts is the oxide formed in the wire or the sample, two considerations we have for this behavior; the shape of the differential conductance - bias voltage shows a typical curve of a tunnel junction [32]. In tunnel junctions the zero bias minimum in the curves is displaced out of the origin. Aspect quite different to a metallic contact junction, where the minimum in the differential conductance always occurs at zero bias voltage [32]. However another important consideration is the value of the differential resistance at zero bias, in our tunnel junctions, this is of the order about 20 - 50 Ω . Several try outs were performed until reproducible data was obtained. Junctions with differential resistances at zero bias voltage between 20–50 Ω gives the most reproducible data. The two crystallographic directions of single crystal were measured with the prepared tunnel junctions; one used to measure

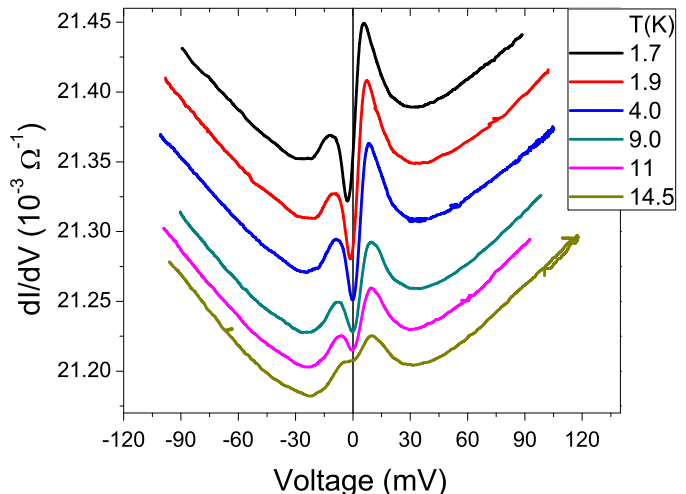


FIG. 1. (Color online) Differential conductance as a function of voltage of URu_2Si_2 -Au(W) tunnel junction, at temperatures from 1.7 K to 14 K. The differential resistance at zero bias is about $\sim 46\text{ }\Omega$. Note the sharp characteristics and the smoothing as the temperature rises. The parabolic background at high bias is typical of a normal tunnel junction. Only the characteristic between $\pm 30\text{ mV}$ presents the Fano resonance, the curves were displaced vertically for better clarity.

over the edge of the crystal axis, the a direction, and the another on the plane of the platelet c axis. For the measurements and characterization of the superconducting state, point contact junctions were used. These were formed with the single crystal and a fine tungsten-gold tip with diameter on the tip, smaller than $5\text{ }\mu\text{m}$, those junctions were prepared at room temperature and glued to a glass substrate with Oxford varnish. The area of the junctions were estimated to be $\sim 1\text{ }\mu\text{m}^2$.

The differential resistance dV/dI as a function of the bias voltage V of the point contacts was measured with the standard modulation ac lock-in amplifier technique, and bridge. The temperature range for characterization of the superconducting state was from 0.325 to about 3 K. For characterization of the hidden order the temperatures range was from 1.7 K to 40 K using a MPMS system from Quantum Design as cryostat, whereas below 2 K a ^3He Oxford refrigerator was used.

The characterization of the work regime of the point contacts was estimated with the Wexler's interpolation formula [33]; we substituted the mean free electronic path $l_i \simeq 100\text{ }\text{\AA}$ [31], the resistivity $\rho \sim 40\text{ }\mu\Omega\text{-cm}$ measured at 2 K [34] and the resistance of the point contacts measured at zero bias voltage. The obtained radii values, between 320 \AA and 3700 \AA , indicate that the contacts are in the diffuse regime, but not far to the ballistic [35].

An important aspect of the junctions used was the thermal stability. In this work we report studies performed in more than 20 different contact junctions, all shown similar $dV/dI(V)$ curves.

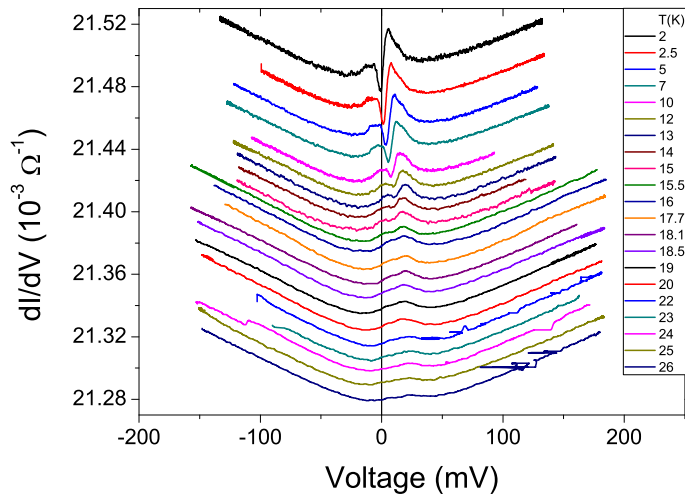


FIG. 2. (Color online) Differential conductance of a tunnel junction in the c direction taken from 2 to 26 K. Note the reduction of the predominant sharp characteristics close to the Fermi level when the temperature is increased. The minimum is displaced from the origin to positive bias voltage. Raising the temperature the structure is smoothed. At high bias voltages the background of the tunnel junction looks normal. All data was vertically displaced for a clear view.

III. RESULTS AND DISCUSSION

Temperature evolution of the differential conductance versus bias voltage from 1.7 to 14.5 K are presented in Fig. 1. These curves reveal the spectroscopic features of the Fano and Kondo resonances, measured in the c crystalline direction. The spectroscopic features of the Fermi surface and the density of states are strongly modified by the Fano and Kondo lattices. Note that in a Peierls distortion the effects are quite different.

The evolution of the spectroscopic features, when temperature is increased, decreases the distortion in the differential conductance. Only close to $T = 17.5$ K where the hidden order develops the distortion is very small. Fig. 2 shows measures performed from 1.7 K to 26 K, in c direction of the crystal, the distortion decreases as the temperature increases, but it persists at high temperature, the curves measured of the differential conductance characteristic at 1.7 K show a typical Fano resonance shape, with very sharp structure. Those characteristics were also observed with similar detail by Elgazzar, et al. and Aynajian, et al. [22, 36]. One set of measurements in the same direction, c , up to 40 K, shown in Fig. 3, displays the typical parabolic background of a normal tunnel junction, with the conductance minimum shifted from the zero bias voltage, out of the origin [37]. However, close examination of the curves still indicates that some features persist at bias voltages at 25–45 mV. Those features may be related to the Kondo lattice.

Fig. 3 shows the smoothing of the central peak and the

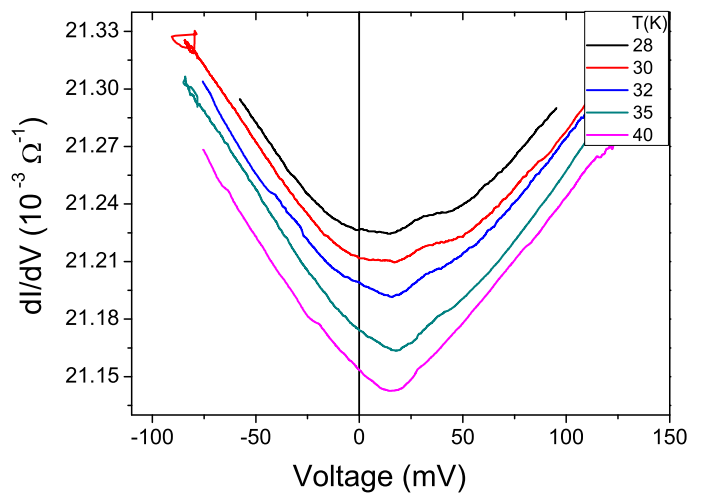


FIG. 3. (Color online) Data for another tunnel junction at high temperature from 28 to 40 K. Here also it is possible to see that small distortion still persist. The minimum observed in the figure 2 now clearly is at about 18 mV. However the distortion of the Fano resonance is difficult to see, at 40 K the characteristic is of a typical normal tunnel junction, if some anomalies persist are difficult to see.

displacement of the minimum out of the origin. However, also the normal parabolic shape of a tunnel junction is noted, but with very small structure [37] perhaps because the remanence of the Kondo lattice. Summarizing, Figure 2 and 3 shows the effect of temperature on the features around zero bias that we are attributed to the formation of a possible arising of a hybridization gap by the Kondo lattice at temperatures for above 70 K [38]. In Figure 4 we show data measured in the a crystallographical direction, these data measured from 20 to 2 K. The spectra are different to the measurements in c direction, in the a direction the differential conductance shows two peaks at ± 10 mV, which tends to decrease as the temperature was increased. At 20 K there is no distortion. Note that the Fano anomaly looks completely different to the observed in the c direction. The data was measured and was reproducible when measured in different junctions. This Fano distortion is different to as the Fano theoretical model [39]. Figure 4 shows two peaks in the differential conductance, these are quite symmetrical with similar magnitude. Those peaks observed at 2 K, are separated from the origin at ± 10 mV, we may consider that the difference of voltage between the peaks may be the size of an energy "gap" of the HO, big, as the gapping in c direction [7]. Similarly as in other figures and directions, figure 4 shows a decreasing structure, tending to be smoothed as temperature is increased. This structure disappears at 20 K. We noted that in a direction no structure exist at 20 K, but in c direction still remains. This is in agreement with the observations by Haule and Kotliar in U or Si site [1]. Also we have to mention that in total accord with Park *et al.* [38] the new features of the

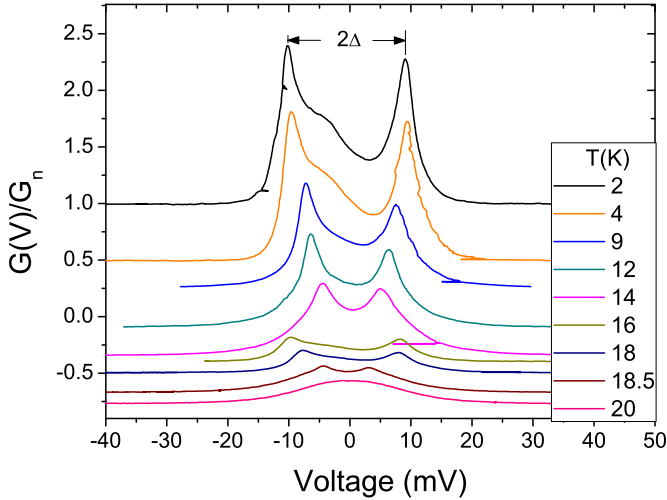


FIG. 4. (Color online) Normalized differential conductance of a tunnel junction measured in the a crystallographic direction. These curves show the presence of a bigger distortion, different to the observed in c crystallographic direction. Note that the structure of the Fano resonance is distorted with two symmetrical peaks at ± 10 mV. The features decreases as the temperature rises, at 14 K the feature look similar to a contact junction with a Z parameter about 0.2 [42].

Fano resonance, as observed in Fig. 4 with measurements taken in the a crystallographic direction of our sample, clearly may indicate another type of a Fano resonance and/or feature of a hybridization gap.

Our experiments, show that nesting depends on the crystallographic direction, and this may be related to size of the HO gap. Measurements in a direction show that $2\Delta = 20$ mV, which is small in c , as already determined by Aynajian *et al.* as ~ 8 mV [22]. With the analysis of the characteristics at temperatures from 40 to 1.7 K, measured in the a direction is possible to do a determination of the HO gap; at 2 K the two maxima are ± 10 mV, and the ratio $2\Delta/K_B T_C$ is ~ 13 .

Point contact experiments reported by Naidyuk *et al.* [40, 41], from temperatures above the superconducting transition up to 20 K, show the differential conductance as a function of bias voltage measured in the c direction and perpendicular to it. In those experiments the features observed in the c direction are not show, meanwhile in the a direction an energy gap is observed with a value about 10 mV, but no features were observed above 17.5 K. Our experiments reported here show features up to 40 K. As mentioned above this behavior over 17.5 K might be interpreted as the arising of a hybridization gap by normalized bands [38]. Our measurements, in the c direction, display more structures which may be associated to an energy gap, persisting above 17.5 K up to 40 K in agreement to Park *et al.* [38].

Studies with PCS were performed at low temperature in the superconducting state to see the evolution with temperature of the energy gap. The features were char-

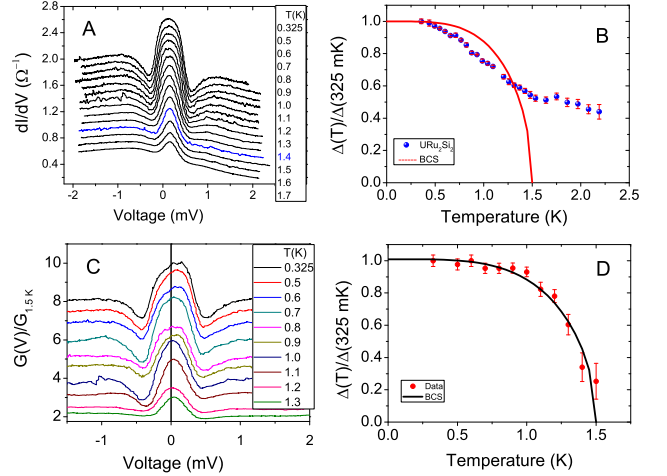


FIG. 5. (Color online) Normalized differential conductance of a point contact in the superconducting state; $\text{URu}_2\text{Si}_2 - \text{Au(W)}$. Panel A shows the rough data measured from 1.7 to 0.3 K. The gap value versus temperature is displayed in B. The evolution does not follow the BCS theory. In order to see the influence of the Fano resonance we subtracted from these data the curve measured at 1.7 K, the resultant curves are shown in panel C, there we plotted the differential conductance normalized at 1.5 K. In D is plotted the superconducting energy gap as a function of normalized temperature, this data follow quite well the BCS theory. This strong and different evolution with temperature of the gap is due to the Fano resonance.

acterized from 3 K to 0.325 K using a ^3He as cryostat. In Figure 5A we show the differential conductance between 0.325 K and 1.7 K, all curves were displaced in the vertical direction by a small amount to have a clear view and details. At temperatures above the superconducting state the Fano resonance may be observed as a distortion at the central part of the curves at zero bias voltage, this behavior was observed and analyzed when plotting the evolution of the gap with temperature. At the lower accessible temperature the superconducting gap feature is well formed, in panel B of figure 5 we display and plot temperature evolution of the gap.

As show in Fig. 5B the energy gap evolution does not follows the BCS model (see continuous line). This anomalous behavior [20] is the effect of the Fano and Kondo resonances. In Fig. 5C it is presented the normalized differential conductance of the same PCS with the structure related to the Fano resonance subtracted in all data. The subtracted data was the differential conductance measured at 1.7 K. The data subtraction was performed in a similar manner as performed by Aynajian *et al.* [22]. Fig. 5 panel D presents the results of this procedure. Data of the superconducting gap as a function of temperature now clearly follow BCS. The characteristics shown in Fig. 5B and 5D, show the influence of the resonances in the evolution with temperature of the gap. Our measurements also indicate that the ratio

$2\Delta/K_B T_C$ is ~ 7.86 - 7.57 . with the transition temperature between 1.35 - 1.4 K, this value is characteristic of a strong coupling superconductor. However, it is very important to mention that recent studies by Kawasaki *et al.*, and Schemm *et al.* [43, 44] have shown time reversal symmetry breaking, using different experimental techniques. Kawasaki, *et al.*, studied the HO and superconductivity using muon spin relaxation spectroscopy, whereas Schemm, *et al.* using Kerr effect. Both experiments found evidence of broken time reversal symmetry, which has a strong implication for both phenomena occurring in URu₂Si₂. Some features below the superconducting state, performed with polar Kerr experiments were observed and as suggested by Schemm *et al.* this effects may imply other physical processes in the superconducting state, and well above in the HO phase at 25 K [44]. Therefore, as mentioned by Kawasaki *et al.* and Schemm *et al.*, those observations may imply a novel pairing mechanism for the superconducting behavior.

IV. CONCLUSIONS

In summary, we have studied the electronic characteristics of URu₂Si₂ with tunnel and point contacts spectroscopies. The spectra show a gap feature with distortions because Fano and Kondo resonances, at high and low temperatures. At low temperature the superconducting gap has an anomalous evolution with temperature. From 0.3 to 2.4 K, we found the influence of the Fano and Kondo resonances and were subtracted, then energy gap follows BCS. From 1.7 to 40 K we observed the presence of the resonances that distorted the features on the Fermi surface, those look different in different crystallographic directions. Lastly, in the *c* direction the structure in the differential conductance persists up to 40 K, whereas in the *a* direction it persists up to 20 K. The HO effects are different in both directions of the Fermi surface.

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- [1] Haule K and Kotliar G 2009 *Nature Phys.* **5** 796
 - [2] Chatterjee S, Trinckauf J, Hänke T, Shai D E, Harter J W, Williams T J, Luke G M, Shen K M, and Geck J 2013 *Phys. Rev. Lett.* **110** 186401
 - [3] Buyers W J L 1996 *Physica B* **223&224** 9
 - [4] Chandra P, Coleman P, Mydosh J A, Tripathi V 2002 *Nature* **417** 831
 - [5] Oh Y S, Kim K H, Sharma P A, Harrison N, Amitsuka H, and Mydosh J A 2007 *Phys. Rev. Lett.* **98** 016401
 - [6] Mydosh J A, Oppeneer P M 2011 *Rev. Mod. Phys.* **83** 1301
 - [7] Maple M B, Chen J W, Dalichaouch Y, Kohara T, Rossel C, Torikachvili M S, McElfresh M W, and J. D. Thompson J D 1986 *Phys. Rev. Lett.* **56** 185
 - [8] Ramirez A P, Coleman P, Chandra P, Bruck E, Menovsky A A, Fisk Z, and Bucher E 1992 *Phys. Rev. Lett.* **68** 2680
 - [9] Ohkuni H, Ishida T, Inada Y, Haga Y Yamamoto E, Onuki Y, and Takahashi S 1997 *J. Phys. Soc. Jpn.* **66** 945
 - [10] Kwok W K, DeLong L E, Crabtree G W, Hinks D G, and Joynt R 1990 *Phys. Rev. B* **41** 11649
 - [11] Motoyama G, Nishioka T, and Sato N K 2003 *Phys. Rev. Lett.* **90** 166402
 - [12] Hassinger E, Knebel G, Izawa K, Lejay P, Salce B, and Flouquet J 2008 *Phys. Rev. B* **77** 115117
 - [13] Broholm C, Lin H, Matthews P T, Mason T E, Buyers W J L, Collins M F, Menovsky A A, Mydosh J A, Kjems J K 1991 *Phys. Rev. B* **43** 12809
 - [14] Palstra T T M, Menovsky A A, and Mydosh J A 1986 *Phys. Rev. B* **33** 6527
 - [15] de Visser A, Kayzel F E, Menovsky A A, Franse J J M, van den Berg J, and Nieuwenhuys G J 1986 *Phys. Rev. B* **34** 8168
 - [16] Walter U, Loong C-K, Loewenhaupt M, and Schlätz W 1986 *Phys. Rev. B* **33** 7875
 - [17] Mason T E and Buyers W J L 1991 *Phys. Rev. B* **43** 11471
 - [18] Schmidt A R, Hamidian M H, Wahl P, Meier F, Balatsky A V, Garrett J D, Williams T J, Luke G M, and Davis J C 2010 *Nature* **465** 570
 - [19] Escudero R, Morales F, and Lejay P 1994 *Phys. Rev. B* **49** 15271
 - [20] Morales F and Escudero R 2009 *J. Low Temp. Phys.* **154** 68
 - [21] Schlätz W, Baumann J, Pollit B, Rauchsvalbe U, Mayer H M, Ahlheim U, and Bredl C D 1986 *Z Phys. B* **62** 171
 - [22] Aynajian P, da Silva Neto E H, Parker C V, Huang Y, Pasupathy A, Mydosh J, and Yazdani A 2010 *Proc. Natl. Acad. Sci.* **107** 10383
 - [23] Villaume A, Bourdarot F, Hassinger E, Raymond S, Taoufour V, Aoki D, and Flouquet J 2008 *Phys. Rev. B* **78** 012504
 - [24] Chandra P, Coleman P, Mydosh J A, and Tripathi V 2002 *Nature* **417** 831

- [25] Rodrigo J G, Guinea F, Vieira S, and Aliev F G 1997 *Phys. Rev. B* **55** 14318
- [26] Giaever I 1960 *Phys. Rev. Lett* **5** 147
- [27] Giaever I and Megerle K 1961 *Phys. Rev. Lett.* **5** 1101
- [28] Yanson I K, Kulik, I O and Batrak A G 1981 *J. Low Temp. Phys.* **42** 527
- [29] Duif A M, Jansen A G M and Wyder P 1989 *J. Phys.: Condens. Matter.* **1** 3157
- [30] Jansen A G M, van Gelder A P and Wyder P 1980 *J. Phys. C: Solid St. Phys.* **13** 6063
- [31] Hasselbach K, Kirtley J R, and Lejay P 1992 *Phys. Rev. B.* **46** 5826
- [32] Jönsson-Åkerman B J, Escudero R, Leighton C, Kim S, and Schuller I K 2000 *Appl. Phys. Lett.* **77** 1870
- [33] Wexler G 1966 *Proc. Phys. Soc.* **89** 927
- [34] Rauchschwalbe U 1987 *Physica B* **147** 1
- [35] Duif A M, Jansen A G M and Wyder P 1989 *J. Phys.: Condens. Matter* **1** 3157
- [36] Elgazzar S, Rusz J, Amft M, Oppeneer P M and Mydosh J A 2009 *Nat. Mat.* **8** 337
- [37] Wolf E L 1989 *Principles of electron tunneling spectroscopy* (Oxford University Press: New York)
- [38] Park W K, Tobash P H, Ronning F, Bauer E D, Sarrao J L, Thompson J D, and Greene L H 2012 *Phys. Rev. Lett.* **108** 246403
- [39] Fano U 1961 *Phys. Rev.* **124** 1866
- [40] Naidyuk Y G, Kvitnitskaya O E, Yanson I K, Nowack A, and Menovsky A A 1995 *Low Temp. Phys.* **21** 236
- [41] Naidyuk y G, Kvitnitskaya O E, Nowack A, Yanson I K, Menovsky A A 1996 *Physica B* **218** 157
- [42] Blonder G E, Tinkham M, and Klapwijk T M 1982 *Phys. Rev. B.* **25** 4515
- [43] Kawasaki I, Watanabe I, Hillier A, and Aoki D 2014 *J. Phys. Soc. Jpn.* **83** 094720
- [44] Schemm E R, Baumbach R E, Tobash P H, Ronning F, Bauer E D, and Kapitulnik A 2014 arXiv: 1410.1479v1